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THE COAGULATION AND DISPERSION OF NON-SPHERICAL AEROSOL PARTICLES IN A TURBULENT ATMOSPHERE

A Final Report

by

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FINAL TECHNICAL REPORT

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UNDER THE SUPERVISION OF PROFESSOR ALAN MATTHEWS
CHAIRMAN OF THE INSTITUTE OF EARTH SCIENCES

Ehud Gavze

I. INTRODUCTION

A. Definitions

1. The Coagulation of particles in the most general sense is a process by which they suffer mutual collisions, which may terminate in coalescence (if they are liquid droplets) or aggregate-formation (if they are solid).

2. The dispersion of Particles is a process by which they occupy increasingly larger portion of space with time.

B. Significance

Both particles coagulation and dispersion are very significant in real life.

As an example, when disseminating fibrous or platelet-like material as an aerosol in the field, the process of coagulation (which mainly occurs in the early, high-concentration stage of the chain of events) will determine particle size and (frequently) complicated shape.

Hence, this process will affect light-scattering properties of the cloud and its deposition-rate on various obstacles in its path; it will also influence dispersivity of the cloud. Aerosol cloud dispersion will determine particle concentration at various space-time points ($\underline{r}$,t) in the field.

C. Mechanisms

1. Coagulation

Aerosol coagulation ensues by several mechanisms which operate in laminar and turbulant flow fields:

i. Brownian coagulation (in laminar and turbulent fields).

ii. Gravitational coagulation (in laminar and turbulent fields), which occurs due to the different fall velocity of the particles.

iii. Gradient coagulation (in laminar and turbulent fields), which occurs due to the passing of particles over each other as they are carried by their stream-lines and thus intercept each other.

iv. Inertial coagulation (in laminar and turbulent fields) which occurs due to the different inertial response of particles to the velocity field of the medium (air), thus causing the emergence of a new relative velocity between the former.

We will be interested in processes within a turbulent atmosphere and in non-spherical particles, which is a completely new research-line.

2. Dispersion

Aerosol dispersion ensues mainly under turbulent conditions; it is caused by the stochastic response of particles to the (turbulent) velocity fluctuations of the medium (air) and thus is a major property of the turbulent field.

The stochastic response depends on particle size, shape and turbulence characteristics.

We will be interested in the stochastic response of non-spherical particles to a turbulent field, which has been only scarcely studied, since coagulation may occur only when the cloud of aerosols is concentrated.

II. PREVIOUS STUDIES

We denote by n_i, $i=1,2,3$ the concentration of single, double, triple particles etc., formed by the process of coagulation.

The Collision Coefficient C_{ij} is defined by the relation:

$$\dot{Q}_{ij} = C_{ij} \cdot n_i \cdot n_j \qquad [1]$$

Q_{ij} is the collision rate of particles i and j with concentrations n_i and n_j.

The concentrations n_k satisfy the following set of equations:

$$\frac{dn_k}{dt} = \frac{1}{2}\sum_{i+j=k} C_{ij}\, n_i n_j - n_k \sum_{j=1}^{\infty} C_{jk}\, n_j \qquad [2]$$

A. Brownian Coagulation

Smoluchowski (1) in his pioneering work on Brownian Coagulation in a still medium assumed that $C_{ij}=C$. The solution to the set of equations [2] under this assumption is:

$$n_k = \frac{n_0 (C n_0 t)^{k-1}}{(1+C n_0 t)^{k+1}} \quad ; \quad n = \frac{n_0}{1+C n_0 t} \qquad [3]$$

Where n_0 is the initial concentration of identical particles and n is the total number of particles. Eq. [2] was later solved numerically without the assumption of equal coeficients.

B. Turbulent Coagulation of Spherical Particles

Here three cases have been treated differently.

1. Asymptotic Case A

Spherical particles which are much smaller than Kolmogoroff's length scale η

$$\eta = \left(\nu^3/\epsilon\right)^{1/4} \qquad [4]$$

ν - kinematic viscosity of medium (air) 0.15 cm^2/sec

ϵ - energy dessipation rate per unit mass 5-200 cm^2/sec^3 in the lower atmosphere.

In the lower atmosphere η = 0.1-0.5 cm

Saffman & Turner (2) obtained a formula for C_{ij} which takes into account the three mechanisms : the inertial response of particles to the flow of air, gravitation and gradients of the

velocity of the air.

$$C_{ij} = 2(2\pi)^{1/2} R_{ij}^2 \Big[(1-\rho/\rho_p)^2 (\tau_i - \tau_j)^2 \overline{\left(\frac{Du}{Dt}\right)^2} + \\ + \frac{1}{3}(1-\rho/\rho_p)^2 (\tau_i - \tau_j)^2 g^2 + \frac{1}{9} R_{ij}^2 \epsilon/\nu \Big]^{1/2} \qquad [5]$$

Here $R_{ij} = r_i + r_j$, r_i being the radius of the i-th particles formed by i original particles.

ρ - is the density of air

ρ_p - density of particles

$\tau_i = 2r_i^2 \rho_p / 9\mu$ - the relaxation time of a Stokesian particle.

μ - dynamic viscosity

g - gravitational acceleration

$\overline{\left(\frac{Du}{Dt}\right)^2}$ - mean square of total time derivative of the air velocity.

This last quantity may be expressed in terms of known quantities of the flow according to Batchelor (3):

$$\overline{\left(\frac{Du}{Dt}\right)^2} = 1.3\left(\epsilon^3/\nu\right)^{1/2}$$

If gravitational and inertial effects are neglected, the expression obtained is:

$$C_{ij} = 1.3 \cdot R_{ij}^3 \, (\epsilon/\nu)^{1/2} \qquad [6]$$

(It differs by a small factor from the expression obtained from [5] upon substituting $\rho = \rho_p$ and $\tau_i = \tau_j$ due to approximations made in deriving it). If one assumes that C_{ij} = 2C one obtains for the densities the same solutions as in Brownian

coagulation, with the characteristics of the turbulent field entering into the constant C.

2. Asymptotic case B

Spherical particles which are much larger than Kolmogoroff's length scale. Abrahamson (4) obtained the following formula:

$$C_{ij} = \frac{5}{16}(r_i + r_j)^2 (v_i'^2 + v_j'^2)^{1/2} \qquad [7]$$

v' is the r.m.s. velocity of the particle. It is related to the r.m.s. velocity of the fluid u' by

$$v' = u'(1 + 1.5\tau\epsilon/u'^2)^{-1/2} \qquad [8]$$

Equation [7] is valid for

$$r^2 > 60\nu u'^2 (\rho/\rho_p)/\epsilon$$

3. Intermadiate Case C

This is the case for which particle's sizes are between the sizes dealt with in cases A and B . A theory for spherical particles, belonging to this case has been developed by Williams & Crane (5).

III. PRESENT STUDY

A. Aim of Study

Inspite of the fact that most real aerosol systems are composed of non-spherical particles, very little has been done so far with respect to the investigation of simultanuous coagulation and dispersion of such particles. This study is consecutive to

those of Krushkal & Gallily (6),(7) on particles orientation which were supported by CRDC, U.S. Army.

As pointed out in the research proposal the aim of this study was to produce a method which would be able to predict

a) particle shape and size distribution and

b) Particle dispersion,

both under the influence of a sufficiently characterized turbulence. This study was limited to the case of particles much smaller than the Kolmogoroff size.

Simultanuous dispersion and coagulation of, initially monodispersed, elongated ellipsoides in a simultated turbulent flow have been calculated up to the stage of two particles collision. The original aim of the study was to calculate also collisions of a single particle with an aggregate and too take account of the hydrodynamic interaction between particles. Due to the death of the chief investigator, Prof. Isaiah Gallily, these goals have not been achieved.

B. General Method

The coagulation-dispersion problem was treated by a Monte Carlo technique in which an ensemble of non-spherical particles was subjected to the effects of a fluctuating velocity and fluctuating velocity gradients in a turbulent air Due to the fluctuating factors, the particles suffered a fluctuating translation and rotation which caused them to coagulate and disperse.

The particles were assumed to be uniformly-sized prolate

ellipsoids of revolution of a large aspect ratio. They were also taken to be much smaller than the typical Kolmagoroff eddy size $(\nu^3/\epsilon)^{1/4}$.

In the Monte Carlo calculations, the tragectories of the particles were calculated from their equation of motion. The inertial memory of the parti les was taken into asccount through their mass and moment of inertia

$$m\frac{d\bar{V}}{dt} = m\bar{g} + \mu\bar{\bar{K}}\cdot(\bar{u}-\bar{V}) \qquad [9]$$

$$I_1\dot{\omega}_1 - (I_2 - I_3)\,\omega_2\omega_3 = L_1 \quad etc. \qquad [10]$$

where $\bar{\bar{K}}$ and $\bar{L}$ on the r.h.s. of these equation are the Oberbeck and Jeffery's sölutions given in (8), (9) and (10).

$$K_{11} = K_{33} = 16\pi a(\beta^2-1) \Big/ \left\{ \frac{2\beta^2-3}{(\beta^2-1)^2} \left(\ln\left[\beta + (\beta^2-1)^{1/2}\right] + \beta \right) \right\} \qquad [11]$$

$$K_{22} = 8\pi a(\beta^2-1) \Big/ \left\{ \frac{2\beta^2-1}{(\beta^2-1)^{1/2}} \left(\ln\left[\beta + (\beta^2-1)^{1/2}\right] - \beta \right) \right\} \qquad [12]$$

$$L_1 = \frac{16\pi\mu\, a\cdot b\cdot c}{3(b^2\beta_0 + c^2\gamma_0)} \left[(b^2-c^2)f' + (b^2+c^2)(\xi' - \omega_1) \right] \qquad [13]$$

u, v are fluid's and particles velocity respectively, a,b,c, are the semi-axes of the ellipsoidal particles.

b>a=c, β=b/a

$$\beta_0 = a \cdot b \cdot c \cdot \int_0^\infty \frac{d\lambda}{(b^2+\lambda)\Delta} \quad ; \quad \gamma_0 = a \cdot b \cdot c \cdot \int_0^\infty \frac{d\lambda}{(c^2+\lambda)\Delta}$$

$$\Delta = \left[(a^2+\lambda)(b^2+\lambda)(c^2+\lambda) \right]^{1/2}$$

f is a strain tensor component

$$f = \frac{1}{2}\left(\frac{\partial w}{\partial y} + \frac{\partial v}{\partial z}\right)$$

ξ is a vorticity component.

$$\xi = \left(\frac{\partial w}{\partial y} - \frac{\partial v}{\partial z}\right)$$

u, v, w, f and ξ are expressed in a system of coordinates attached to the ellipsoid and parallel to its principal axes. It was assumed that during each time interval of particle motion, the velocity gradient was kept constant, though it changed from one interval to another according to a probability function, truncated around its zero mean.

IV. RESULTS

A. Turbulent field properties

For the turbulent field we took the following values:

$$\varepsilon = 10^3 \text{cm}^2/\text{sec}^3, \quad \nu = 0.15\text{cm}^2/\text{sec}.$$

Thus for the typical Kolmogoroff length we had:

$$\eta = 4.3 \cdot 10^{-2} \text{cm}$$

The Kolmogoroff typical velocity was 3.5 cm/sec and the typical time-$1.2 \cdot 10^{-2}$ sec.

The velocity gradients within each eddy were assumed to be normally random and constant during a time interval Δt_i. The velocity $\underline{u}$ is given by

$$\underline{u}(\underline{x}+d\underline{x}) = \underline{u}(\underline{x}) + \frac{\partial \underline{u}(\underline{x})}{\partial x_j} dx_j \qquad [14]$$

The normal distribution of the velocity gradients was truncated at $\pm 4\delta$, δ given by the following relations between the gradients and typical values of the turbulent field.

$$\overline{\frac{\partial u_i}{\partial x_k}\frac{\partial u_j}{\partial x_l}} = \begin{cases} \varepsilon/15 & (i=j=k=l) \\ -\varepsilon/30 & (i=k=j=l \text{ or } k=j=l=i) \\ 2\varepsilon/15 & (k=l=i=j) \\ 0 & (\text{all other combinations}) \end{cases} \qquad [15]$$

B. Method of Calculations

The particles were given randomly distributed initial translational and rotational velocities with zero mean. Since the relaxation time of the particles was much shorter than a time interval, the particles' translational velocity was, practically, the velocity of the air. the rotational velocity was calculated, at each time step, from equation [13]. At the end of each time interval $t=10^{-3}$ sec., new gradients were generated at the location of the particle according to [15].

The new translational velocity of the particle was calculated according to [14] and the new rotational velocity according to [13] and [10], with the velocity gradients, appearing in [13], taken at the new point. During each time interval collisions were checked at intervals of 10^{-4} sec. in 2-D calculations and $2 \cdot 10^{-5}$ sec. in 3-D calculations.

C. 2-D Calculations

Calculations were performed first in two dimensions in order to check the method.

The particles were prolate ellipsoids of revolution with semi axes b and a, chosen according to a log normal distribution.

The geometrical mean length - $\langle 2b \rangle = 2 \cdot 10^{-3}$ cm

g.r.m.s. $\delta_g = 1.2 \cdot \langle 2b \rangle$

The geometrical mean width - $\langle 2a \rangle = 2 \cdot 10^{-4}$ cm

g.r.m.s. $\delta_g = 1.2 \cdot \langle 2a \rangle$

Truncation was at ± 0.15 of the mean size

Particles density $\rho_p = 1.\ g/cm^3$

The time intervals were

$$\Delta t_i = \begin{cases} 10^{-4}\,\text{sec} & \text{for collision checking} \\ 10^{-3}\,\text{sec} & \text{for constant velocity gradients} \end{cases}$$

Total time of calculation : $\Sigma \Delta t_i = 3 \cdot 10^{-2}$ sec.

Initial inter-particle distance $= 10^{-2}$ cm.

Initial particle concentration $= 10^4/cm^3$

Initial number of particles $= 100.$

Fig. 1 and Fig. 2 show two typical numerical experiments. In these experiments the particles were separated after each collision and the coagulation continued. In each of the experiments the number of particles was 64. In the first one (Fig. 1) 27 collisions were counted, in the second - 12.

D. 3-D Calculations

As in the 2-D calculations the particles were prolate ellipsoids of revolution of log-normally distributed sizes. We aimed at obtaining relations between the size and the initial concentration of the particles and the number of collisions. Therefore calculations were performed with two sets of particles' sizes and concentrations.

Sizes

i. regular particles

mean length - $\langle 2b \rangle = 4 \cdot 10^{-4}$ cm

g.r.m.s. $\delta_g = 1.2 \cdot \langle 2b \rangle$

mean width - $\langle 2a \rangle = 0.8 \cdot 10^{-4}$ cm

g.r.m.s. $\delta_g = 1.2 \cdot \langle 2a \rangle$

ii. large particles

All linear dimensions are twice as large.

Truncation was at ± 0.15 of the mean size.

Concentrations

i. regular concentration

initial number of particles	N= 125
inter particle initial distance	$X= 10^{-3}$ cm
initial number concentration	$n_0 = 10^9\ cm^{-3}$

ii. high concentration

initial number of particles	N= 125
inter particle initial distance	$X= 5 \cdot 10^{-4}$ cm
initial number concentration	$n_0 = 8 \cdot 10^9\ cm^{-3}$

In addition the effect of the initial number of particles was checked.

Initial number

i. N=64

ii. N=125

particles density $\rho_p = 1 g/cm^3$

time intervals

$$\Delta t_i = \begin{cases} 2 \cdot 10^{-5} \text{ sec} & \text{for collision checking} \\ 10^{-3} \text{ sec} & \text{for constant velocity gradients} \end{cases}$$

Total time of calculation : $\Sigma \Delta t_i = 2 \cdot 10^{-2}$ sec.

1. Dispersion

A typical picture of the dispersion of the particles is shown in Fig. 3. It shows that after $2 \cdot 10^{-2}$ sec the particles are so dispersed that it is unlikely to expect many more collisions to

occur.

2. Collisions

The number of collisions depends non-linarily on the initial concentration of the particles. According to Saffman & Turner (2) the number concentration of pairs, as a function of time is:

$$n_2 = \frac{n_0^2 \tau}{(1+n_0\tau)^3} \quad ; \quad \tau = 5.2 \cdot R^3 (\epsilon/\nu)^{1/2} t \qquad [16]$$

R is the radius of initial spheres.

For $n_0\tau \ll 1$

$$n_2 \simeq n_0^2 \tau \qquad [17]$$

In our calculations $(\epsilon/\nu)^{1/2} = 1.2 \cdot 10^{-2}\,sec^{-1}$. The values of τ/t, based on half the length and half the width of the paeticle and on the initial number concentration $n_0 = 8 \cdot 10^9\,cm^{-3}$, are:

a. For $R=4 \cdot 10^{-5}\,cm$ $\quad \tau/t=4 \cdot 10^{-15}$ [18]

b. For $R=2 \cdot 10^{-4}\,cm$ $\quad \tau/t=5 \cdot 10^{-13}$ [19]

In Fig. 4 typical results of numerical experiments are shown with particles of size (i), number concentration $n_0 = 8 \cdot 10^9\,cm^{-3}$ and initial number N=125. It shows that most collisions occur in the first $3 \cdot 10^{-4}$ sec and that the total number of collisions after this time is 30-40. According to [17] and [18] the time needed for this number of collisions is ~ 10^{-4} sec and according to [19] ~ 10^{-6} sec. The longer time needed in our calculations is due to the strong dispersion which was not accounted for in (2).

In Fig. 5 similar results are shown for larger particles of size (ii) and initial concentration $n_0 = 10^9 cm^{-3}$.

3. Effect of Concentration

In Fig. 6 the number of collisions is given for three initial concentrations (the graph shows the average over great number of numerical experiments). The lowest concentration - $n_0 = 1.25 \cdot 10^8 cm^{-3}$ seems to exhibit a too low number of collisions for any conclusion to be drawn from it. The average slope a_1, corresponding to initial concentration $n_0 = 10^9 cm^{-3}$ in the first $2 \cdot 10^{-4}$ sec, is $a_1 \simeq 5/2$ whereas the slope a_2, corresponding to $n_0 = 8 \cdot 10^9 cm^{-3}$ is $a_2 \simeq 25/2$ so that $a_2/a_1 = 5$.

Similar results are shown in Fig 7. Here the initial number of particles was N=64. The ratio of the slopes of the two graphs taken at $t = 1.4 \cdot 10^{-4}$ sec is $a_2/a_1 = 6.25$. It should be noted that the leveling off in the number of collisions for N=125 occurs after $t = 2 \cdot 10^{-4}$ sec whereas for N=64 after $t = 1.4 \cdot 10^{-4}$ sec. This is due to the lower number of particles. These results are in disagreement with [16] and [17], from which a ratio of $a_2/a_1 = 64$ is to be expected. This disagreement is a consequence of the small number of particles introduced initially in the experiment and of the rapid dispersion shown above. Nevertheless, the strong dependence of the number of collisions on the initial concentration is still obvious.

4. Effect of Size

Larger particles are heavier but the mass does not enter into the expression [16]. Also it does not enter in our calculations since the relaxation time was small compared to the time step. Therefore inertial effects were not significant in the range of sizes we have applied. Thus, the only parameter that determines the results, if the initial number of particles N is large enough, is the ratio of the spacing between particles and their sizes. An increase by a factor of 2 of each linear dimension of a particle should therefore be equivalent to an increase by a factor 8 of the initial concentration. In Fig. 8 the number of collisions is shown for regular particles of size (i) and for large particles of size (ii). The initial number is N=125. In Fig. 9 comparison is made between regular particles of size (i) with initial concentration $n_0=8\cdot10^9 cm^{-3}$ and between larger particles (ii) and initial concentration $n_0=10^9 cm^{-3}$. The similarity between the results assures us of the validity of the method of calculations.

5. Effect of Initial Number

Our calculations were performed with a small number of particles due to computational limitations. The increase of the initial number, i.e. the volume of equal concentration of interacting particles, results in an increase in the number of collisions. This is roughly explained by the fact that if two equal volumes of particles are put together, particles from the two volumes will interact through the common surface. Thus, the number of collisions will be greater than the sum of collisions

in the two separated volumes. The effect of increase in N is shown in Fig. 10. It should be noted that not only the number of collisions increased with the increase of N, but also the time interval at which high rate of collisions occur is larger. Thus, with N=64 the time at which the graph of collisions levels off was $t \simeq 1.4 \cdot 10^{-4}$ sec whereas for N=125 $t \simeq 1.4 \cdot 10^{-4}$ sec.

V. CONCLUSION

A numerical model was constructed for the simulation of dispersion and coagulation of a cloud of non-spherical aerosol particles in turbulent conditions. Coagulation was observed in the early stages of the dispersion of the cloud. It appears from the calculations that, in a small cloud, dispersion is the dominant process from the early stages of the formation of the cloud and coagulation stops after a short time. Nevertheless, at the time coagulation occured it exhibited a behaviour similar to what is expected from existing theories but modified by the strong dispersion of the cloud. The intention of the chief investigator, Prof. I. Gallily, was to perform numerical simulations with a much larger number of particles, to examine the behaviour of the system with particles of different aspect ratios and to take account of the particle-particle hydrodynamic interaction. Due to the death of Prof. Gallily these additional goals were not achieved.

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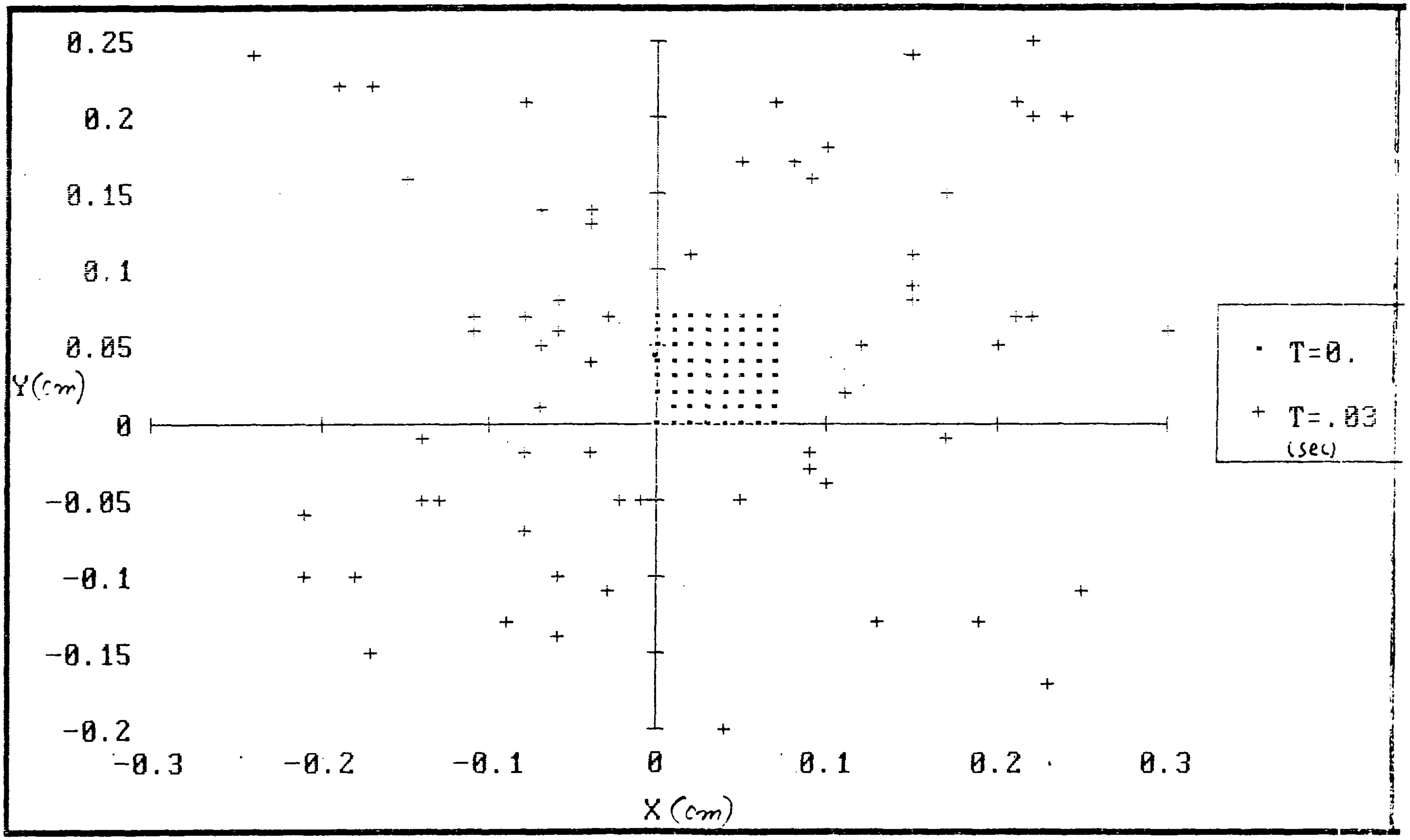

Fig. 1 2-D Dispersion

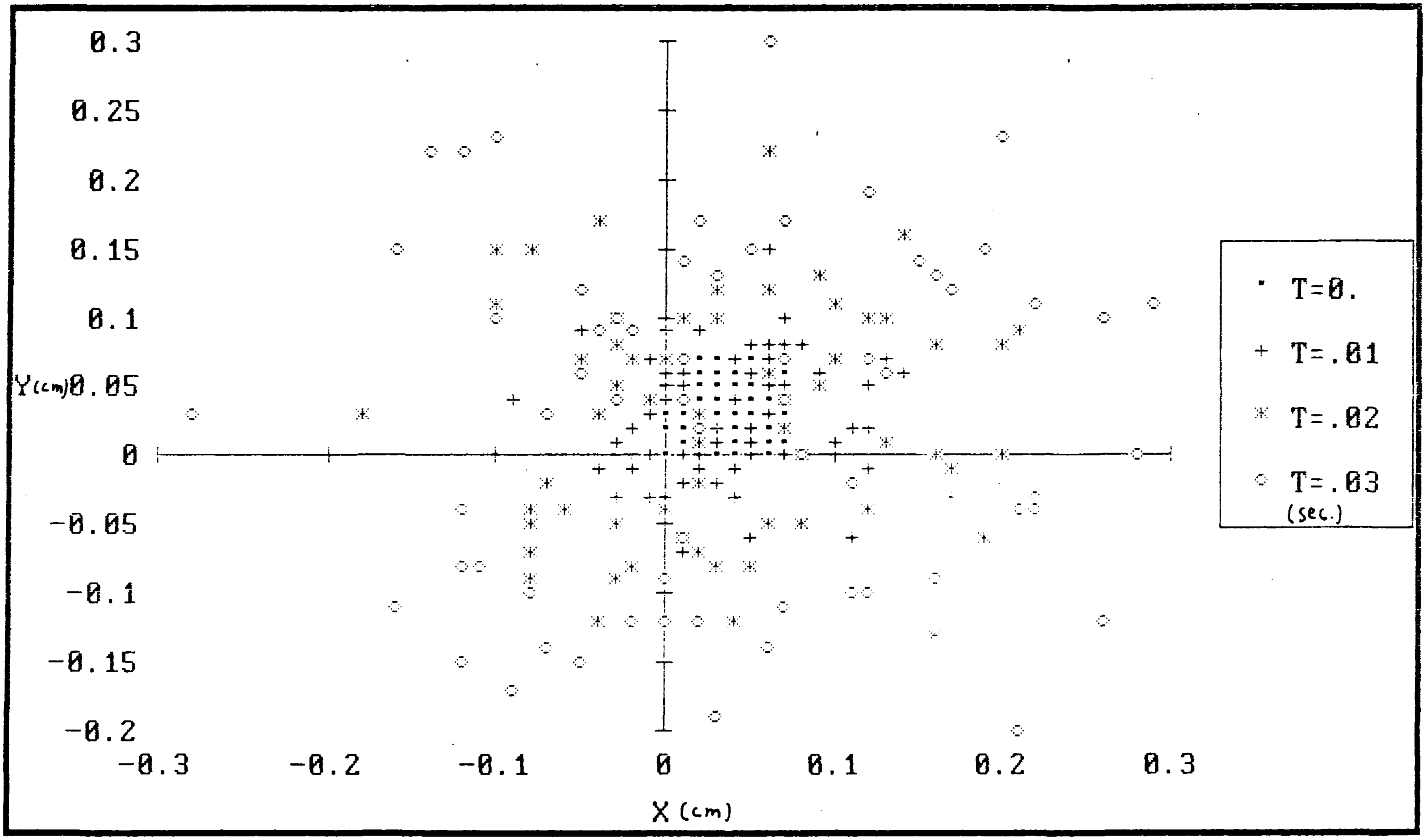

Fig. 2 2-D Dispersion

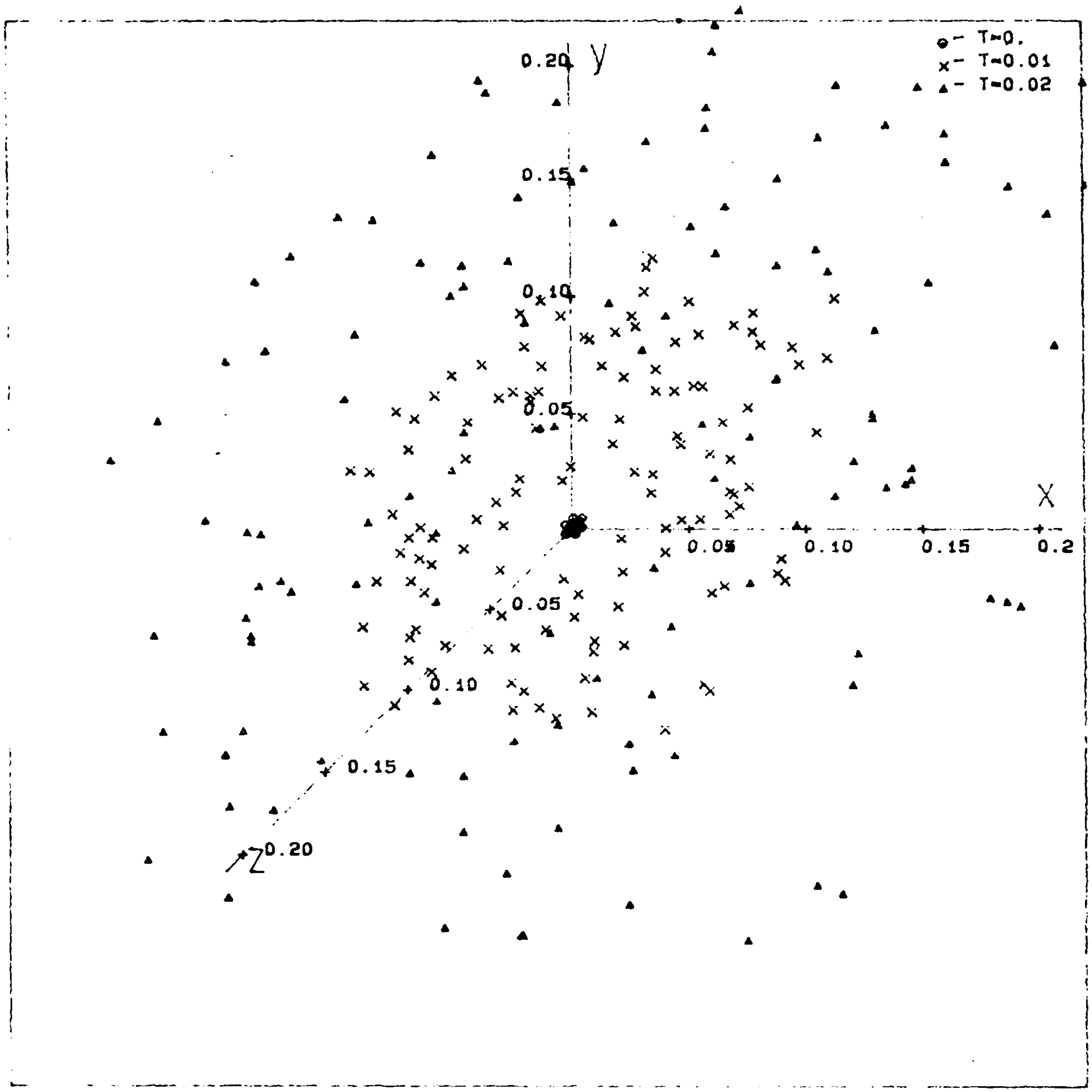

Fig. 3 - 3-D Dispersion

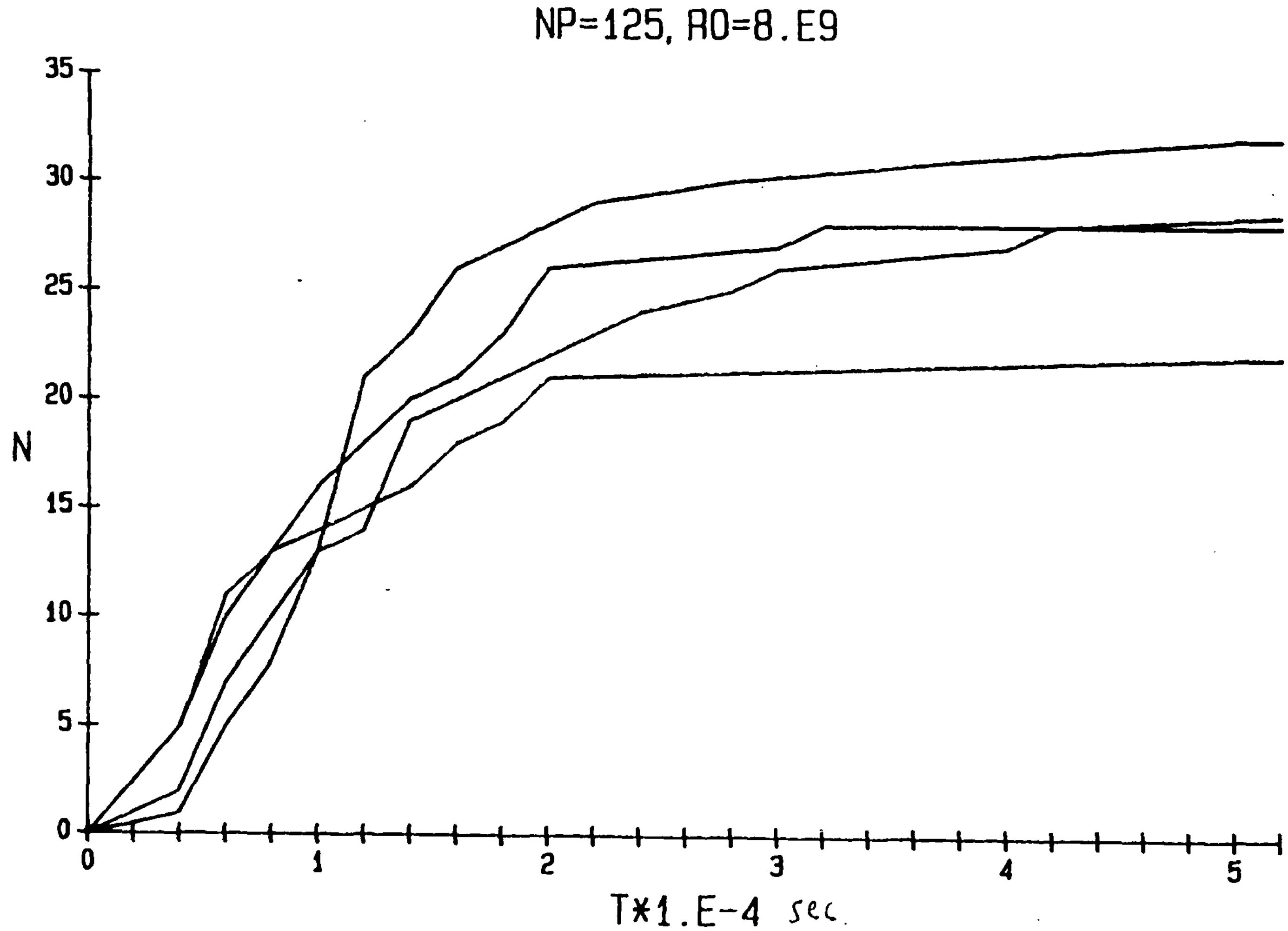

Fig. 4 - Number of collisions

particle size (i), initial concentration (ii)

NP=initial number, RO=initial number concentration

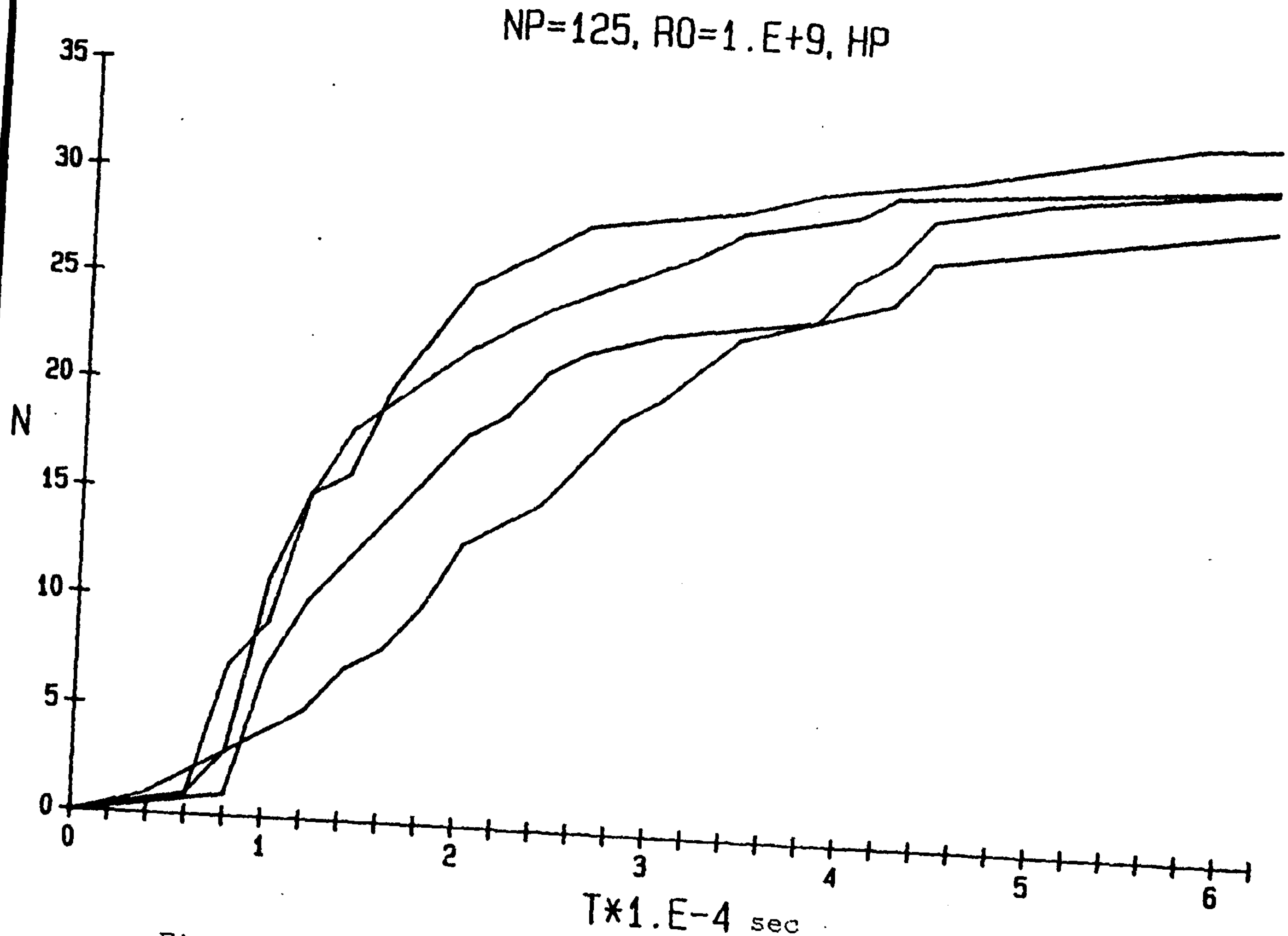

Fig. 5 - Number of collisions

particle size (ii), initial concentration (i)

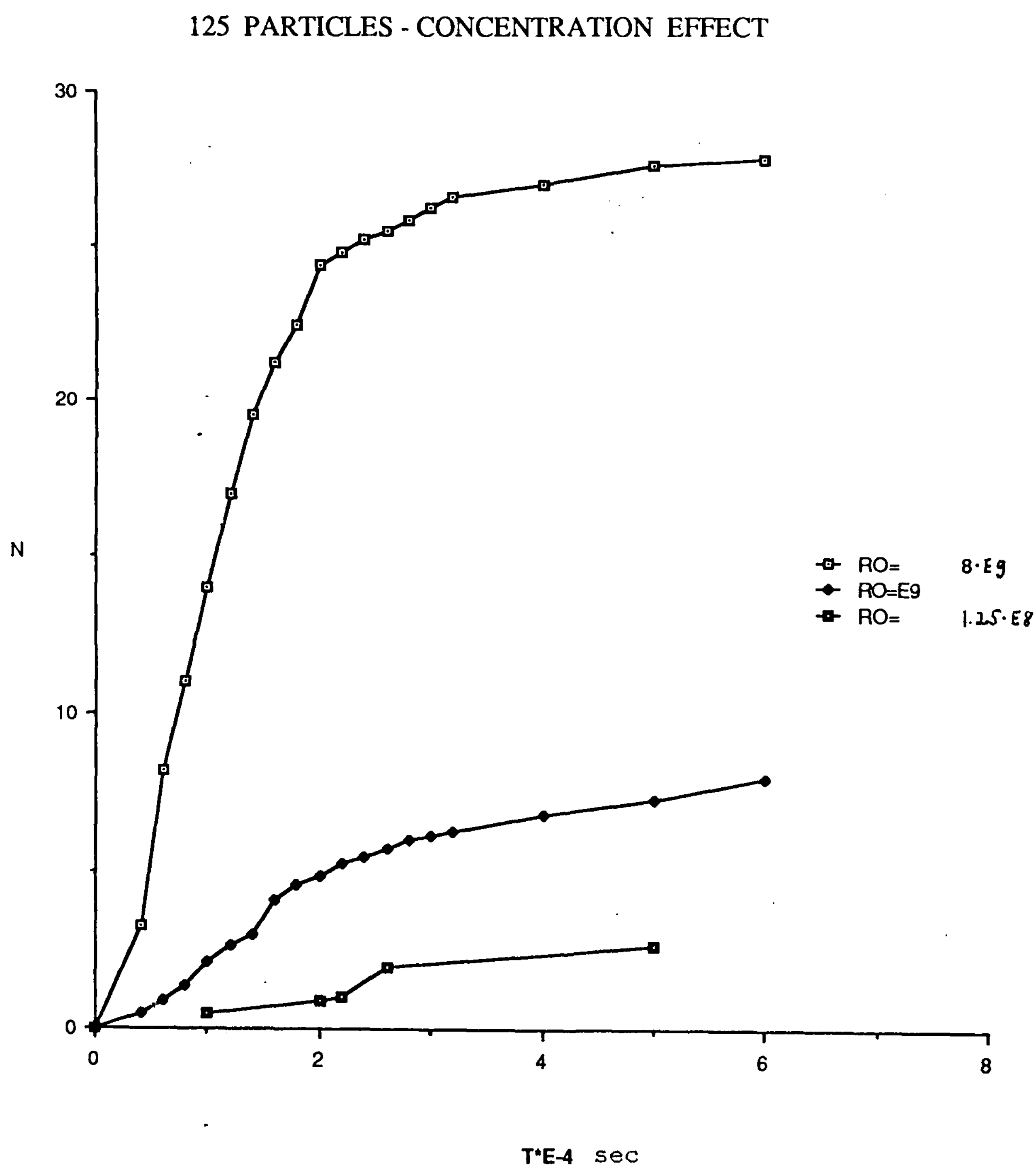

Fig. 6 - Number of collisions for 3 initial concentrations RO particle size (i), initial number NP=125

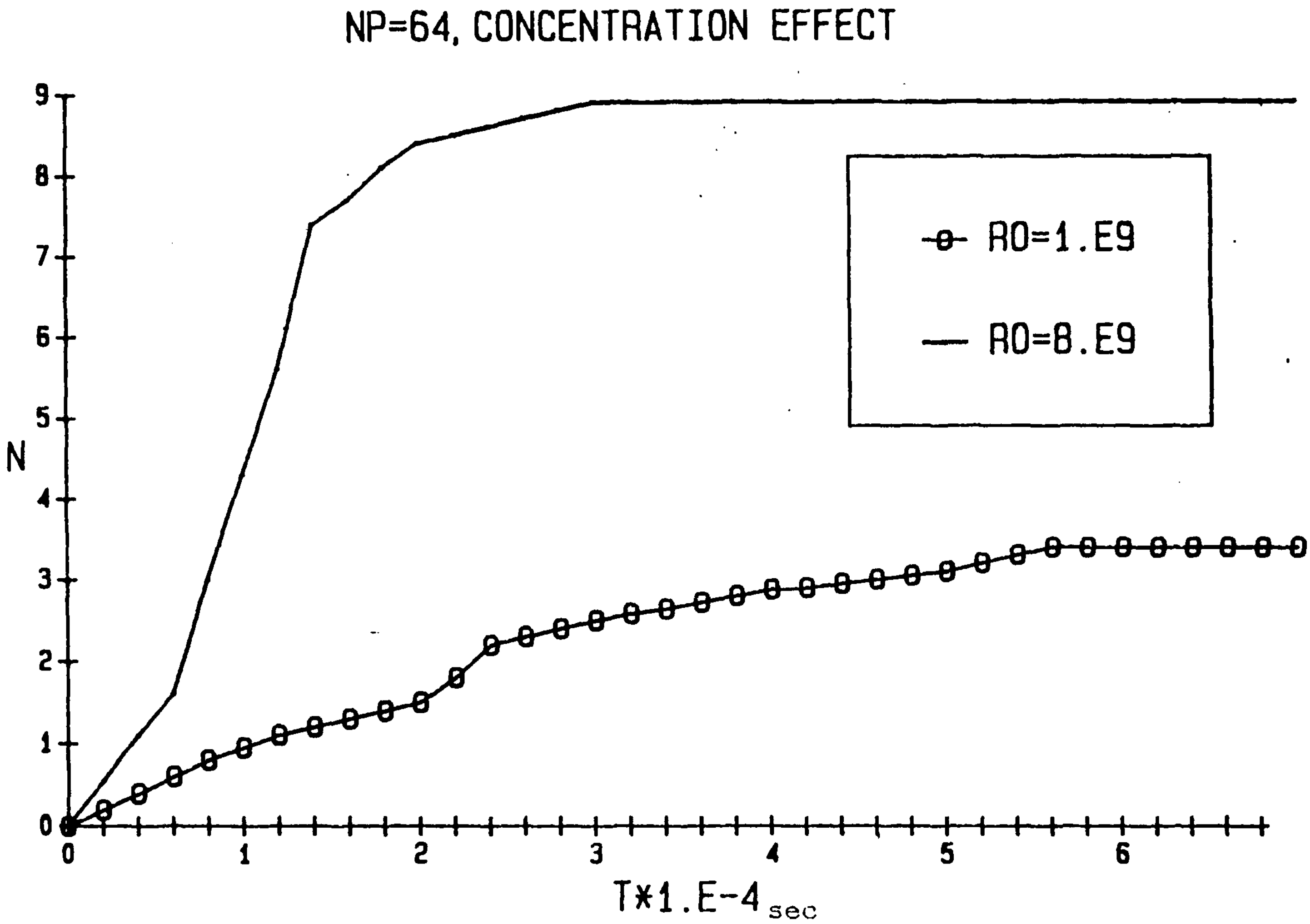

Fig. 7 - Number of collisions for 2 initial concentrations RO particle size (i), initial number NP=64

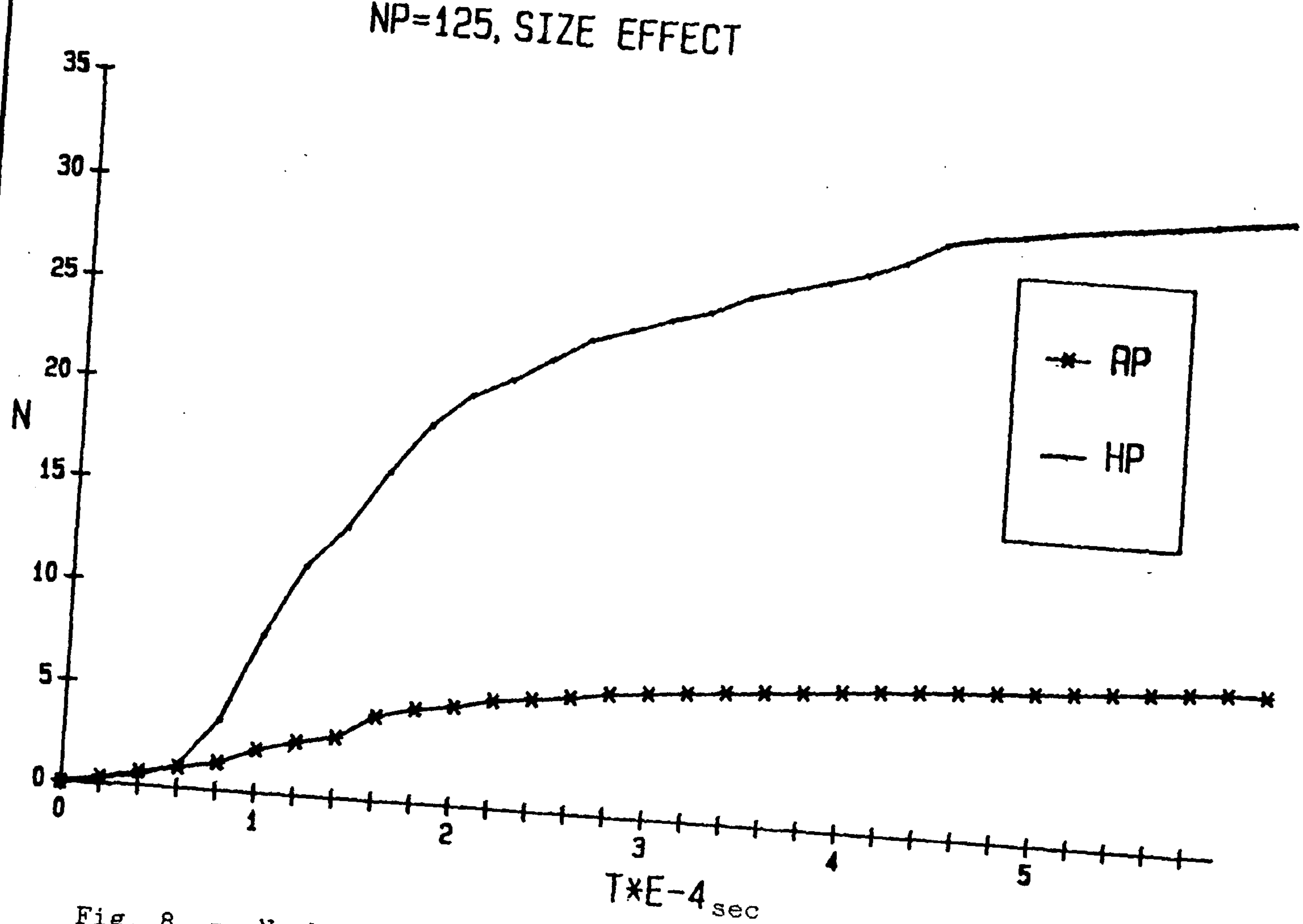

Fig. 8 - Number of collisions
RP - particle of size (i), HP - particles of size (ii)
initial number NP=64

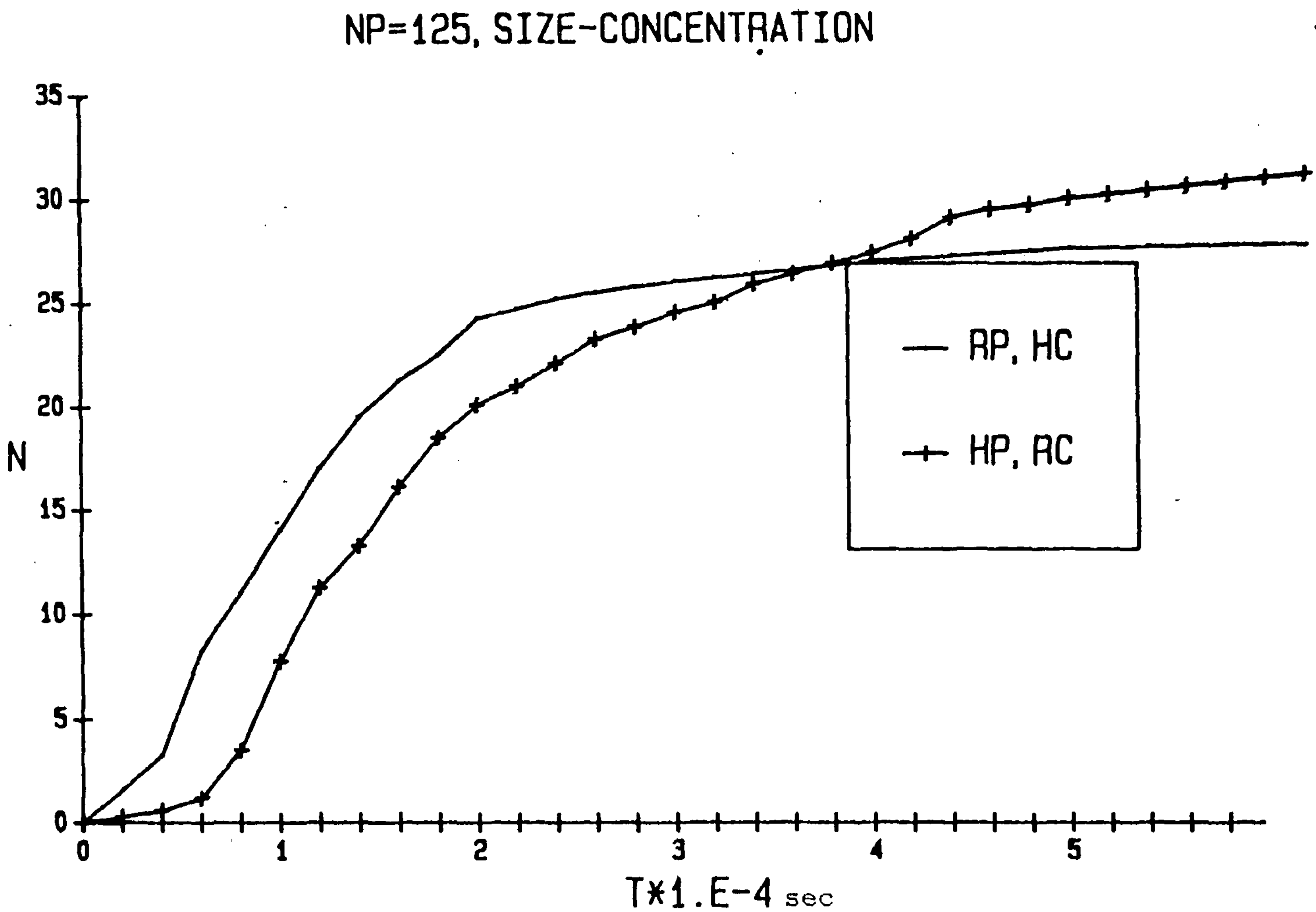

Fig. 9 - Number of collisions
RP,HC - particles of size (i) initial concentration (ii)
HP,RC - particles of size (ii) initial concentration (i)

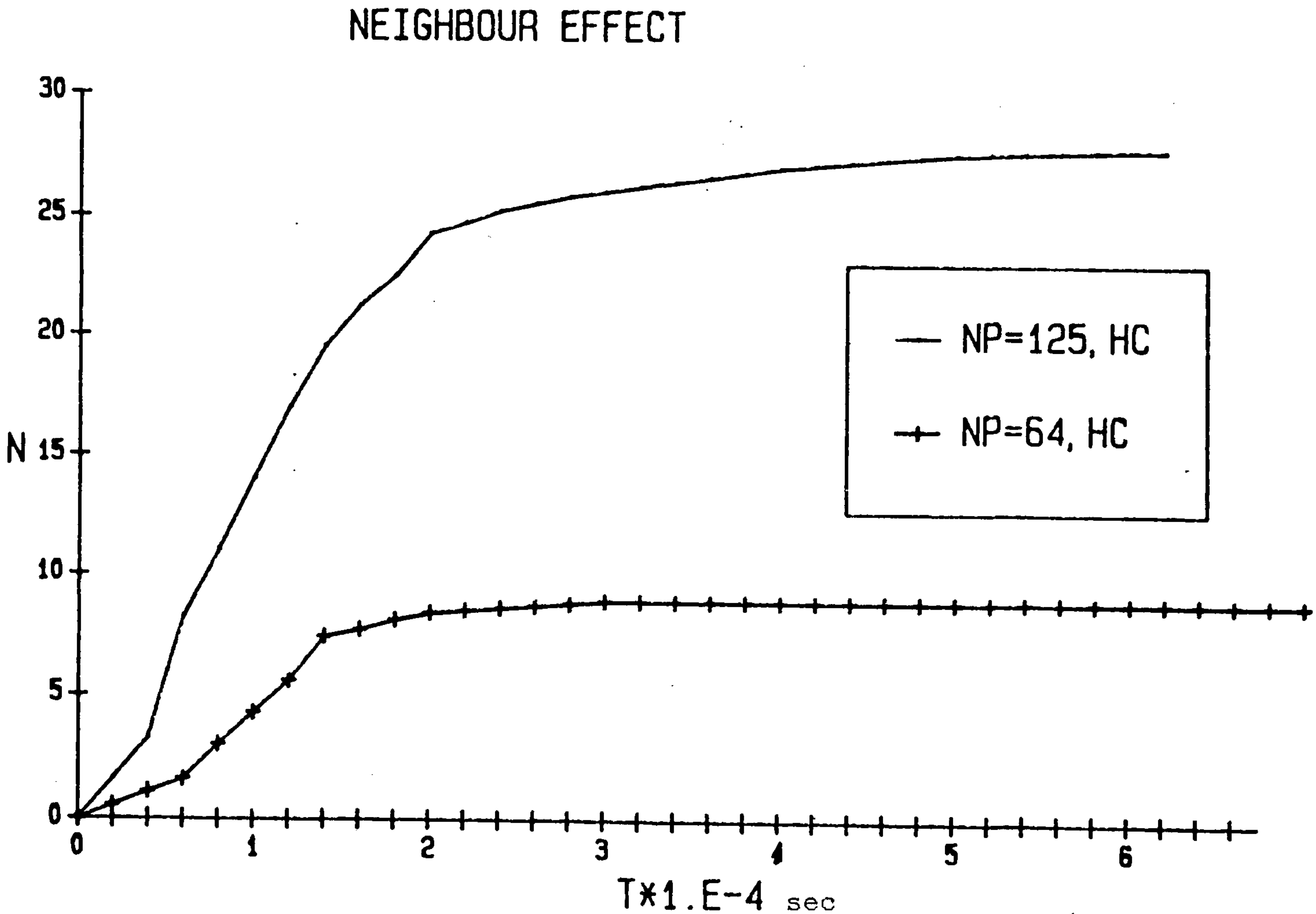

Fig. 10 - Number of collisions
NP - initial number of particles
HC - concentration (ii), particle size (i).